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## **JAPANESE / ENGLISH TRANSLATION OF**

**Japanese Patent Application JP 2001 - 354842 A**

**Polyester Composition for Calendering**

**Your Ref: 103003 - 08**

**For: Eastman Chemical Company**



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(Continued on last page)

(54) [Title of the Invention]

**Polyester Composition for Calendering**

(57) [Summary]

**[Object]** A polyester composition for calendering with significantly reduced tackiness and adhesion to rolls during polyester calendering, and with the ability to yield polyester sheets or films of excellent transparency.

**[Means of Achievement]** The composition is produced by means of melt mixing 0.001 to 10 parts by mass of polyolefin wax (B) per 100 parts by weight of copolyester (A) comprising terephthalic acid units and ethylene glycol units as the main units, and also comprising other



copolymer units at 5 mol% or more of all the structural units. Good calenderability that compares favorably with that of vinyl chloride or EVA can be achieved.

**[Claims]**

**[Claim 1]** A polyester composition for calendering, characterized in that 0.001 to 10 parts by mass of polyolefin wax (B) are melted and mixed with 100 parts by mass of copolyester (A) comprising terephthalic acid units and ethylene glycol units as the main components thereof, and also comprising other copolymer units at 5 mol% or more of all the structural units.

**[Claim 2]** The polyester composition for calendering according to claim 1, characterized in that the number-average molecular weight of polyolefin wax (B) is 50,000 or less.

**[Claim 3]** The polyester composition for calendering according to claim 1 or 2, characterized in that polyolefin wax (B) is modified polyolefin wax.

**[Claim 4]** The polyester composition for calendering of claim 3, characterized in that polyolefin wax (B) is modified polyethylene wax.

**[Claim 5]** The polyester composition for calendering of claim 1, characterized in that copolyester (A) is a polyester block copolymer comprising carboxylic acid monomer units (a) that contain 80 to 100 mol% of aromatic dicarboxylic acid units, and glycol monomer units (b) that contain 0.5 to 15 mol% of polyalkylene glycol units with a number-average molecular weight of 500 to 3,000, wherein

the combined content of the dibasic acid units other than terephthalic acid units in the carboxylic acid monomer units (a), and the dibasic acid units other than ethylene glycol units in glycol monomer units (b) is 20 mol% or greater;

polycarboxylic acid units with three or more carboxyl groups are further contained in the carboxylic acid monomer units (a), and/or polyhydric alcohol units with three or more hydroxyl radicals are further contained in the glycol monomer units (b), with the combined content of the polycarboxylic acid units with three or more carboxyl groups and the polyhydric alcohol units with three or more hydroxyl radicals being 0.05 to 2 mol%; and

the intrinsic viscosity  $[\eta]$ , as measured in a mixed solvent of equal amounts of phenol/tetrachloroethane at 25°C, is 0.6 to 1.2 dL/g.



## **[Detailed Description of the Invention]**

**[0001]**

**[Technological Field of the Invention]** The present invention relates to a polyester composition which has excellent calenderability and with which sheets or films with excellent transparency can be molded.

**[0002]**

**[Prior Art]** Polyethylene terephthalate and other polyester resins have various excellent properties, including transparency, mechanical properties, and gas barrier properties. These resins are also advantageous from a sanitation and safety standpoint in that there is little concern over residual monomers or harmful additives when the resins are molded. Therefore, their use has spread to industrial materials, including sheets, in place of the vinyl chloride resins that have been widely used in the past. The method whereby a molten thermoplastic resin is extruded through flat dies and brought to the desired thickness as a result of being passed through polishing rolls is a popular method for molding sheets from polyester resin. Calendering is widely used as a method of molding sheets from resins. Such calendering is a method whereby the resin is held in the space between calender rolls at a high temperature and plasticized, defoamed, kneaded, and rolled to the desired thickness between the rolls. Its advantages include that sheets can be mass-produced with good thickness accuracy and the like. Calendering is often used, principally for vinyl chloride, ethylene-vinyl acetate copolymer (EVA), and rubber.

**[0003]** Nevertheless, ordinary polyester resin is not suitable for calendering, and is therefore almost never calendered. This is because polyester inherently is very tacky, to such an extent that it is often used as a hot melt adhesive, and there is a problem when it is calendered in that it is too tacky and adheres the rolls. Moreover, there is a tendency toward an even further increase in tackiness when the roll temperature is raised to facilitate plasticizing and defoaming. Even though there is a slight reduction in tackiness when the roll temperature is lowered, polyester is a crystalline resin with an obvious melting point. Therefore, the resin suddenly becomes incapable of being plasticized when the roll temperature is reduced from the optimal temperature near the melting point, and it is very difficult to release and take off a stable film from the rolls. Consequently, polyester is not industrially calendered today.



[0004] Moreover, blending of vinyl chloride graft polymer with polyester is disclosed only in JP (Kohoku) 58-56545 as technology whereby calenderability of polyester elastomer is improved, and very few conventional examples are known. In addition, when the polymer blend composition in JP (Kohoku) 58-56545 is used in order to improve the calenderability of a polyester elastomer, it is difficult to maintain the original excellent properties (such as oil resistance, heat resistance, and mechanical properties) of the polyester elastomer because the oil resistance and heat resistance of the vinyl chloride graft polymer blended with the polyester elastomer are inferior to those of the polyester elastomer, and, therefore, a satisfactory product cannot be obtained. When calenderability modifiers (metallic soaps and aliphatic amides), which are often used with actual results in resins for calendering, are used with polyester elastomers, there is either no improvement of calenderability or marked blooming is produced. Furthermore, there is virtually no improvement in calenderability when a silicone oil disclosed in JP (Kokai) 61-195154 or an inorganic hard fine powder such as talc disclosed in JP (Kokai) 58-129046 is added as a slidability improver of polyester elastomer, or when the montanic acid ester wax disclosed in JP (Kokai) 57-8238 is added as an extrusion moldability improver of polyester elastomers in order to make it easier for the elastomers to be released from rolls. Admixing a specific additive into polyester elastomers is proposed in JP (Kokai) 7-278418 as a technology for improving calenderability of polyester elastomers. Nevertheless, there are virtually no reported examples of techniques in which the calenderability of polyester resin is improved for the purpose of obtaining highly transparent sheets and films.

[0005] The present invention is intended to solve the above-mentioned problems, its purpose being to provide a polyester composition for calendering that possesses significantly reduced tackiness and adhesion to rolls during polyester calendering, and can yield polyesters sheets or films with excellent transparency.

[0006]

**[Means Used to Solve the Above-Mentioned Problems]** The polyester composition for calendering of the present invention is characterized in that 0.001 to 10 parts by mass of polyolefin wax (B) are melted and mixed with 100 parts by mass of copolyester (A) comprising terephthalic acid units and ethylene glycol units as the main components thereof, and also comprising other copolymer units at 5 mol% or more of all the structural units. Here, polyolefin wax (B) is preferably one with a number-average molecular weight of 50,000 or less. Moreover,



a modified polyolefin wax, particularly a modified polyethylene wax, is preferred as polyolefin wax (B). In addition, copolyester (A) is preferably a polyester block copolymer comprising carboxylic acid monomer units (a) that contain 80 to 100 mol% of aromatic dicarboxylic acid units, and glycol monomer units (b) that contain 0.5 to 15 mol% of polyalkylene glycol units with a number-average molecular weight of 500 to 3,000, wherein the combined content of the dibasic acid units other than terephthalic acid units in the carboxylic acid monomer units (a) and the dibasic acid units other than ethylene glycol units in glycol monomer units (b) is 20 mol% or greater; polycarboxylic acid units with three or more carboxyl groups are further contained in the carboxylic acid monomer units (a), and/or polyhydric alcohol units with three or more hydroxyl radicals are further contained in the glycol monomer units (b), with the combined content of the polycarboxylic acid units with three or more carboxyl groups and the polyhydric alcohol units with three or more hydroxyl radicals being 0.05 to 2 mol%; and the intrinsic viscosity  $[\eta]$ , as measured in a mixed solvent of equal amounts of phenol/tetrachloroethane at 25°C, is 0.6 to 1.2 dL/g.

[0007]

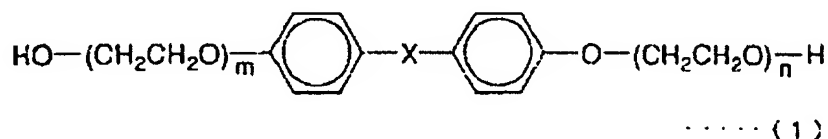
**[Embodiments of the Invention]** The present invention will now be described in detail. The polyester composition of the present invention comprises copolyester (A) and polyolefin wax (B). The copolyester (A) used in the present invention comprises terephthalic acid units and ethylene glycol units as the main components, and also comprises other copolymer units at 5 mol% or more of all the structural units. Specific examples of the dicarboxylic acid component other than terephthalic acid for producing copolyester (A) of the present invention include aromatic dicarboxylic acids such as phthalic acid, isophthalic acid, 2,6-naphthalene dicarboxylic acid, diphenyldicarboxylic acid, and diphenoxyethanedicarboxylic acid; aliphatic dicarboxylic acids such as oxalic acid, succinic acid, glutaric acid, adipic acid, pimelic acid, sebacic acid, azelaic acid, and decanedicarboxylic acid; and alicyclic dicarboxylic acids such as cyclohexanedicarboxylic acid. Of these, isophthalic acid is preferably used because it is inexpensive. Specific examples of diol components other than ethylene glycol that are used to produce the copolyester (A) of the present invention include diethylene glycol, triethylene glycol, propylene glycol, 1,4-butanediol, 1,6-hexanediol, neopentyl glycol, dimer diol,



cyclohexanediol, cyclohexane dimethanol, alcohol components represented by the following general formulas (1), (2), and (3), and ethylene oxide adducts of derivatives thereof.

[0008]

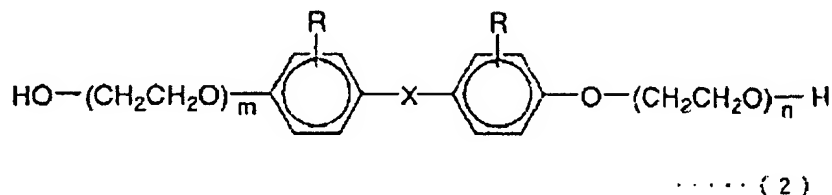
[Chemical formula 1]



(In the formula, X is either CH<sub>2</sub>, C(CH<sub>3</sub>)<sub>2</sub>, C(CF<sub>3</sub>)<sub>2</sub>, O, S, or SO<sub>2</sub>, and n and m satisfy 1 ≤ n + m ≤ 4.)

[0009]

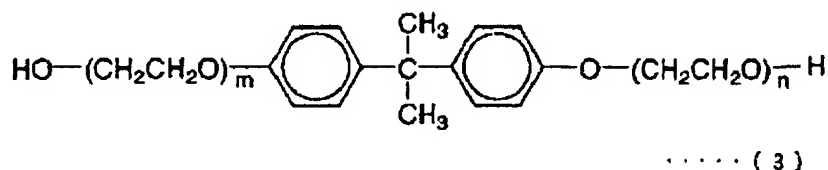
[Chemical formula 2]



(In the formula, X is either CH<sub>2</sub>, C(CH<sub>3</sub>)<sub>2</sub>, C(CF<sub>3</sub>)<sub>2</sub>, O, S, or SO<sub>2</sub>; R is an alkyl group with 1 to 5 carbons; and n and m satisfy 1 ≤ n + m ≤ 4.)

[0010] Of these, the ethylene oxide adduct of bisphenol A having the structure shown by the following general formula (3) is preferred.

[Chemical formula 3]



(In the formula, n and m satisfy 1 ≤ n + m ≤ 4.)

[0011] In order to reduce crystallinity and to guarantee transparency after calendering, copolyester (A) of the present invention comprises, in addition to terephthalic acid units and ethylene glycol units, other copolymer units at 5 mol% or more of all the structural units. Moreover, the copolyester preferably comprises other copolymer units at 15 to 50 mol% of all the structural units because the calendering temperature can be reduced in such a case. However, making the content of copolymer units greater than 50 mol% of all the structural units will lead



to a reduction in mechanical properties and heat resistance of the resulting copolyester, and will also be disadvantageous economically. The other copolymer units are selected from the above-mentioned dicarboxylic acid units and diol units, but it is preferred that 50 mol% or more of the copolymer units be isophthalic acid because it is inexpensive.

[0012] Copolyester (A) is preferably a polyester block copolymer that comprises carboxylic acid monomer units (a) containing 80 to 100 mol% of aromatic dicarboxylic acid units, and glycol monomer units (b) containing 0.5 to 15 mol% of polyalkylene glycol units with a number-average molecular weight of 500 to 3,000, with the combined amount of the dibasic acid units other than terephthalic acid units in the carboxylic acid monomer (a) units and the dibasic acid units other than ethylene glycol units in glycol monomer (b) units being 20 mol% or higher. The heat resistance of the resulting polyester is improved when the polyester comprises 80 mol% or more of aromatic dicarboxylic acids, while there is a reduction in the mechanical strength of the resulting polyester when the aromatic dicarboxylic acids account for less than 80 mol% of the carboxylic acid monomer units (a). Polyethylene glycol, polypropylene glycol, polyethylene glycol-polypropylene glycol block copolymers, polytetramethylene glycol, polyhexamethylene glycol, and the like can be cited as examples of polyalkylene glycol units that can be used as the copolymer components in copolyester (A), and they can be used alone or as a mixture. If the average molecular weight of the copolymer component is less than 500, flexibility will be insufficient, and if it exceeds 3,000, compatibility with the polymer will be poor, polymerization will be slow, and there will be a tendency toward a reduction in the mechanical strength of the molded product. The particularly preferred molecular weight ranges from 800 to 2000. The preferred amount of the polyalkylene glycol used ranges from 0.5 to 15 mol%. If this amount is less than 0.5 mol%, there will be no effect in terms of imparting flexibility, while if the amount exceeds 15 mol%, there will be a reduction in thermal stability, polymerization will be slow, and there will be a tendency toward a reduction in the mechanical properties of the molded product. The particularly preferred amount of polyalkylene glycol is 1 to 10 mol%.

[0013] The combined amount of the dibasic acid units other than terephthalic acid units in the carboxylic acid monomer units (a), and the dibasic acid units other than ethylene glycol units in the glycol monomer units (b) that form the polyester block copolymer should be 20 mol% or higher. If the combined amount is less than 20 mol%, there will be a tendency toward a reduction in the transparency of the polyester over time. It is particularly preferred that the



combined amount be 30 to 50 mol%. If the combined amount is within this range, a polyester with excellent heat resistance and a controlled reduction in transparency over time will be obtained.

[0014] It is preferred that the composition of the present invention contain a polycarboxylic acid with three or more carboxyl groups and/or a polyhydric alcohol compound with three or more hydroxyl radicals as a component of copolyester (A) of the present invention. Even a small amount of a polycarboxylic acid with three or more carboxyl groups and/or a polyhydric alcohol with three or more hydroxyl radicals will effectively endow the resin with melt tensile strength. Polycarboxylic acids such as trimellitic acid, pyromellitic acid, and anhydrides thereof, and polyhydric alcohols such as trimethylolpropane, pentaerythritol, and glycerin can be cited as specific examples. The amount in which the polycarboxylic acid with three or more carboxyl groups and/or the polyhydric alcohol compound with three or more hydroxyl radicals is added should be within a range of 0.05 to 2 mol%, and preferably 0.1 to 1 mol%, of all the structural units. The reaction tends to be more difficult to control due to gelling when this amount exceeds 2 mol%.

[0015] Polyester is usually obtained by means of introducing a dicarboxylic acid component and a diol component into a reactor, raising the temperature by means of heating, performing an esterification or transesterification, and then performing polymerization. A commonly used esterification catalyst or transesterification catalyst, such as titanium butoxide, dibutyltin oxide, magnesium acetate, manganese acetate, or the like, can be used as needed for the esterification or transesterification. The water or alcohol that has been produced as a result of the reaction is removed in accordance with conventional methods after esterification or transesterification. It is preferred that polymerization is performed while the diol component is distilled out under a vacuum of 150 mmHg or less. Conventional catalysts such as titanium butoxide, dibutyltin oxide, tin acetate, zinc acetate, tin disulfide, antimony trioxide, and germanium dioxide can be cited as examples of conventional polymerization catalysts for polymerization. Moreover, there are no special restrictions to the polymerization temperature or the amount of catalyst, and they can be selected as needed. The preferred degree of polymerization corresponds to an intrinsic viscosity  $[\eta]$ , as determined in a mixed solvent of equal amounts of phenol/tetrachloroethane at 25°C, of 0.6 to 1.20 dL/g. The particularly preferred intrinsic viscosity is 0.7 to 1.20 dL/g. If the intrinsic viscosity is less than 0.6 dL/g, the melt viscosity will be low and there will be a



reduction in moldability, making it difficult to obtain a satisfactory sheet or film. Moreover, if the intrinsic viscosity is larger than 1.20 dL/g, there will be a tendency toward poor molded appearance.

[0016] The polyolefin wax (B) that is used in the present invention is preferably a polyolefin with a number-average molecular weight of 50,000 or lower, particularly 1,000 to 30,000.

Methods for producing such polyolefin wax include those whereby an  $\alpha$ -olefin starting material is polymerized by means of the high-pressure method or the low-pressure method, and those whereby the molecular weight is reduced as a result of the decomposition of polyolefin once it has been polymerized to a high molecular weight. If the molecular weight of the polyolefin wax is greater than 50,000, the polyolefin will have poor compatibility with the polyester, and the surface appearance of the molded article will tend to be adversely affected. Furthermore, a so-called modified polyolefin wax, in which carboxylic acid groups and hydroxyl groups are introduced into the molecules by means of oxidation or the like, maleic acid or epoxy group-containing compounds are reacted, or unsaturated groups are added, is preferred because it enhances the effect of improving roll release properties. Ethylene, propylene, 1-butene, 1-hexene, 4-methylpentene, and the like can be cited as examples of the  $\alpha$ -olefins used the starting materials of the polyolefin wax. Two or more  $\alpha$ -olefins can also serve as the starting materials, and two or more types can be jointly used. Of these, polyethylene wax, polypropylene wax, and polybutylene wax with a molecular weight of 30,000 or lower are preferred. Modified polyethylene waxes with a molecular weight of 10,000 or less and an acid value (or hydroxyl group value, as measured by means of the JIS K5902 method) of 10 or higher has a particularly good effect in terms of improving roll release properties, and is therefore preferred. An acid value of 15 to 25 is particularly preferred. If the acid value exceeds 25, there will be a reduction in the effect of improving roll release properties. The amount of polyolefin wax (B) used in the present invention is 0.001 to 10 parts by mass per 100 parts by mass of polyester. The particularly preferred amount is 0.01 to 5 parts by mass. If this amount is less than 0.001 part by mass, there will be an insufficient reduction of tackiness and adhesion to the calender roll, while if this amount exceeds 10 parts by mass, transparency and mechanical properties of the polyester will be compromised.

[0017] There are no special restrictions to the method of adding polyolefin wax (B) to copolyester (A). Examples include methods whereby polyolefin wax is mixed with the polyester



in a melted state immediately after the reaction; methods whereby the wax is added to produced polyester chips, heated, melted, and mixed; methods whereby the wax is dry blended with the polyester chips immediately before calendering; methods whereby the polyolefin wax is introduced into the calendering process; methods whereby the wax is added to the polyester in molten or semi-molten form during calendering; and the like.

[0018] The composition of the present invention may also contain conventional hindered phenols, phosphites, thioethers, and other antioxidants; benzophenone, hindered amines, and other weather resistance agents; thickeners such as epoxy compounds and isocyanate compounds; mold release agents such as silicone oil, stearic acid metal salts, montanic acid metal salts, and montanic acid ester waxes; coloring agents such as dyes and pigments; ultraviolet absorbers such as titanium oxide, carbon black, and the like; reinforcing agents such as glass fibers, carbon fibers, and potassium titanate fibers; fillers such as silica, clay, calcium carbonate, calcium sulfate, and glass beads; and nucleators such as talc; as well as flame retarders, plasticizers, foaming agents, phosphors, antifungal disinfectants, crosslinking agents, surfactants, and the like. These are added as needed as long as their content is within a range that does not compromise the purpose of the present invention. These additives and fillers can be premixed with the polyester, they can be mixed with the polyester together with the composition of the present invention, or they can be added after the composition of the present invention has been prepared.

[0019]

#### [Working Examples]

##### [Example 1]

688.76 g of terephthalic acid, 172.19 g of isophthalic acid, 450.63 g of ethylene glycol, and 3.47 g of trimethylol propane were introduced into a reactor and heated and stirred for three hours at 190 to 225°C to perform esterification. Then the temperature was raised to 260°C, and 0.30 g of antimony trioxide was added as the polymerization catalyst to promote polymerization. Copolyester (A1) was obtained. The composition of polyester (A1) is shown in Table 1. The intrinsic viscosity  $[\eta]$  of this polyester (A1) was 0.74 dL/g. Additive (B1) shown in Table 2 was dry blended with the pellets obtained by means of drying this resin for 36 hours at 60°C, and these pellets were placed between the rolls of a calender with two six-inch rolls (manufactured



by Kansai Roll) and kneaded and molded into sheets by means of calendering. The roll temperature was 190°C. The release properties and transparency of the resulting sheets were evaluated.

**[Examples 2 through 7 and Comparative Examples 1 through 4]**

Other than the fact that each of the additives listed in Table 2 was admixed as shown in Table 3, various polyester compositions or sheets were made and evaluated as described in Working Example 1.

**[0020]**

**[Working Example 8]**

Polyester (A2) was obtained as described in above-mentioned Working Example 1 using 2.8 kg of terephthalic acid, 1.27 kg of ethylene glycol, 0.74 kg of 1,4-cyclohexanedimethol, 6.87 g of trimethylolpropane, 1.25 kg of polytetramethylene glycol with an average molecular weight of 1,000, 62.5 g of tetrakis[methylene(3,5-di-t-butyl-4-hydroxyhydrocinnamate)] methane, 0.15 g of phosphoric acid, and 1.58 g of germanium dioxide. Polyester (A2) had an intrinsic viscosity  $[\eta]$  of 0.76 dL/g. Additive (B1) shown in Table 2 was dry blended with pellets obtained by means of drying this resin for 36 hours at 60°C. Sheets were molded as described in Working Example 1 with the exception that the roll temperature was 160°C. The release properties and transparency of the resulting sheets were evaluated.

**[Working Examples 9 through 11, Comparative Examples 5 through 8]**

Other than the fact that each additive listed in Table 2 was admixed as shown in Table 3, each polyester composition or sheet was produced as described in Working Example 8 and similarly evaluated.

**[0021]** Each property was evaluated as follows.

<Intrinsic viscosity> The intrinsic viscosity was measured (dL/g) in a mixed solvent of equal amounts of phenol/tetrachloroethane at 25°C.

<Calendering tests> Release of the sheet from the roll during sheet molding was evaluated in 5 steps.

"5": Can be released with virtually no resistance and on a level in no way inferior to ordinary vinyl chloride

"4": Release properties are somewhat worse than step "5"

"3": Release properties are so inadequate that mass production is difficult



"2": Release properties are so inadequate that calendering appeared impossible

"1": Could not be released

<Transparency of molded article> Transparency of a 500- $\mu$ m sheet produced by means of calendering was evaluated macroscopically.

"5": Transparent enough that characters and pictures on the other side can be clearly seen through the sheet

"4": Transparency is somewhat lower than step "5"

"3": Cloudy

"2": So cloudy that characters and letters on the other side can be just barely seen through the sheet; not suitable for uses that require transparency

"1": So cloudy that characters and letters on the other side cannot be seen through the sheet

[0022]

[Table 1]

| Copolyester                |                              |           | A1   | A2   |
|----------------------------|------------------------------|-----------|------|------|
| Composition<br>(mol%)      | Dicarboxylic acid units      | TPA       | 80   | 100  |
|                            |                              | IPA       | 20   | 0    |
|                            | Diol units                   | EG        | 100  | 62   |
|                            |                              | CHDM      | 0    | 30   |
|                            |                              | PTMG 1000 | 0    | 7.7  |
|                            | Polyhydric alcohol component | TMP       | 0.5  | 0.3  |
| Intrinsic viscosity (dL/g) |                              |           | 0.74 | 0.76 |

TPA: Terephthalic acid

IPA: Isophthalic acid

EG: Ethylene glycol

CHDM: Cyclohexanedimethanol

PTMG 1000: Polytetramethylene glycol (number-average molecular weight 1,000)

TMP: Trimethylolpropane



[0023]

[Table 2]

| Additive | Additive name                           | Manufacturer (company name)     | Remarks   |
|----------|---|---------------------------------|---|
| B1       | Polyethylene wax                        | Allied Signal (AC-316A)         | Acid value type (acid value: 18)<br>Number-average molecular weight: 5000     |
| B2       | Polyethylene wax                        | Allied Signal (AC-629A)         | Acid value type (acid value: 18)<br>Number-average molecular weight: 1400     |
| B3       | Polyethylene wax                        | Mitsui Chemicals (Hi-Wax 220MP) | Acid value type (acid value: 1)<br>Number-average molecular weight: 2000      |
| B4       | Polyethylene wax                        | Mitsui Chemicals (Hi-Wax 4202E) | Low acid value type (acid value: 17)<br>Number-average molecular weight: 2600 |
| B5       | Polyethylene wax                        | Mitsui Chemicals (Hi-Wax 2203A) | Acid-modified type (acid value: 30)<br>Number-average molecular weight: 2700  |
| B6       | Montanic acid butylene glycol ester wax | Hoechst (Hoechst Wax OP)        |   |
| B7       | Calcium stearate                        |                                 |   |
| B8       | Glycerin monostearate                   | Riken Vitamin (Rikemal S-100A)  |   |

[0024]

[Table 3]

|                       | Polyester | Additive |                              | Sheet release properties | Sheet transparency |
|-----------------------|-----------|----------|------------------------------|--------------------------|--------------------|
|                       |           | Type     | Amount added (parts by mass) |                          |                    |
| Working Example 1     | A1        | B1       | 0.5                          | 5                        | 4                  |
| Working Example 2     | A1        | B1       | 0.25                         | 4                        | 5                  |
| Working Example 3     | A1        | B2       | 1.0                          | 5                        | 1                  |
| Working Example 4     | A1        | B3       | 0.5                          | 5                        | 2                  |
| Working Example 5     | A1        | B3       | 0.25                         | 4                        | 3                  |
| Working Example 6     | A1        | B4       | 0.5                          | 5                        | 2                  |
| Working Example 7     | A1        | B5       | 1.0                          | 3                        | 1                  |
| Working Example 8     | A2        | B1       | 0.5                          | 5                        | 4                  |
| Working Example 9     | A2        | B2       | 1.0                          | 4                        | 2                  |
| Working Example 10    | A2        | B3       | 0.5                          | 3                        | 2                  |
| Working Example 11    | A2        | B4       | 0.5                          | 3                        | 2                  |
| Comparative Example 1 | A1        | B6       | 2.0                          | 1                        | —                  |
| Comparative Example 2 | A1        | B7       | 2.0                          | 1                        | —                  |
| Comparative Example 3 | A1        | B8       | 2.0                          | 1                        | —                  |
| Comparative Example 4 | A1        | —        | —                            | 1                        | —                  |
| Comparative Example 5 | A2        | B6       | 2.0                          | 1                        | —                  |
| Comparative Example 6 | A2        | B7       | 2.0                          | 1                        | —                  |
| Comparative Example 7 | A2        | B8       | 2.0                          | 1                        | —                  |
| Comparative Example 8 | A2        | —        | —                            | 1                        | —                  |



The amounts in which the additives are added are shown in parts by mass, where the amount of polyester is taken as 100 parts by mass.

[0025] It is clear from the results in Table 3 that adding polyethylene wax to the copolyester results in obvious improvement of calenderability, particularly a reduction in the tackiness and adhesion to the rolls, and that the transparency of the resulting sheet can be improved by means of selecting the appropriate type of polyethylene wax.

[0026]

[Effect of Invention] The present invention allows a polyester with good calenderability that compares favorably with that of vinyl chloride or EVA to be obtained by means of melting and mixing a specific compound with the polyester. In particular, tackiness and adhesion to the roll can be considerably reduced and a high-quality transparent polyester sheet or film can be obtained. Particularly good surface appearance of the molded article can be realized as a result of adding a polyolefin wax with a number-average molecular weight of 50,000 or less. Using a modified polyolefin wax as this polyolefin wax enhances the effect of improving the roll release properties. In other words, the use of modified polyethylene wax can enhance considerably the effect of improving the roll release properties.

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